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# Impact of Degraded Al-SNF on Shipping and Basin Storage

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## ABSTRACT

An evaluation of the impact of breached cladding of aluminum-based spent nuclear fuel (Al-SNF) has been completed. The evaluation has focused on the impact of the resulting exposed fuel core material on radioactive material containment during shipping and on the radioactive material release to basin water during wet storage. Degraded fuel assemblies from the RA-3 Research and Test Reactor that experienced corrosion while stored in water basins at the Ezeiza Central Storage Facility (Ezeiza CSF) in Argentina were evaluated as a case study of foreign research reactor fuel being sent to the Savannah River Site (SRS). One hundred thirty-four assemblies of a total of 207 RA-3 assemblies have each been observed to exhibit up to 3 $cm^2$ of exposed fuel meat. The results of the containment analysis show that this fuel can readily meet shipping requirements, without canning and in a standard shipping cask with a standard leak rate less than $2.89 \times 10^{-5}$ atm·$cm^3$/sec. Storage of this aluminum-based fuel would result, at most, in a minor increase in the steady-state activity level of the SRS L-Basin water, well within the operational limit.

## INTRODUCTION

Aluminum-clad nuclear fuel stored in water basins is subject to corrosion attack if the water quality and storage conditions are aggressive to aluminum [1-4]. Spent fuel damaged by such conditions may require canning or other special handling measures to meet containment requirements for shipping, and to avoid excessive release to water of the storage basin that may result in excessive water activity levels.

An inspection of spent fuel from the RA-3 Research and Test Reactor at the Comision Nacional de Energia Atomica (CNEA) Ezeiza Atomic Center near Buenos Aires, Argentina has been completed [5]. The fuel was known to have corrosion damage and the inspection was performed to characterize the damage prior to planning for shipment to the SRS. The results from visual examination at the Ezeiza CSF performed in October 1999 [5-6] showed that 134 of the 207 assemblies had exposed fuel meat with an area estimated at 3 $cm^2$/assembly in the worst case.

Figures 1 and 2 show typical corrosion damage exposing fuel meat on the RA-3 assemblies. Exposed fuel meat surface areas were estimated from photographs such as these. On assembly S-133 shown in Fig. 1, corrosion nodules can be seen in the active region of the fuel plate. Note that each of the nodules appears to have originated in a scratch on the surface of the fuel clad. Figure 2 shows the cladding damage that is apparent with the corrosion product nodules dislodged, exposing the fuel meat.

The impact to shipping and storage considering regulatory requirements and SRS safety authorization bases has been performed for a test case of the RA-3 fuel.

## CONTAINMENT ANALYSIS

A radionuclide inventory was developed for the RA-3 fuel and a containment analysis was performed to determine if the fuel could be shipped as planned in a standard shipping cask with a leak rate of $1.0 \times 10^{-6}$ atm·$cm^3$/sec.

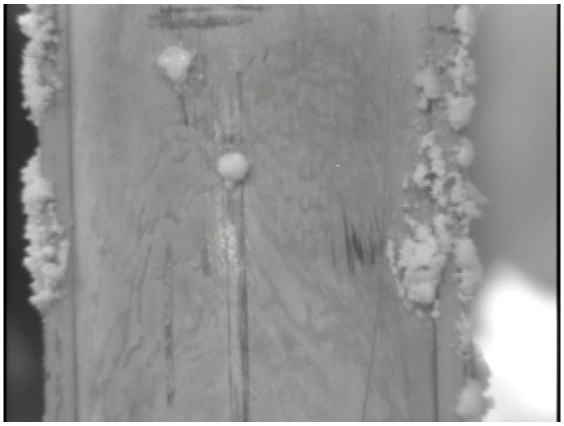

**Figure 1. Corrosion nodules associated with scratches and crevices.**

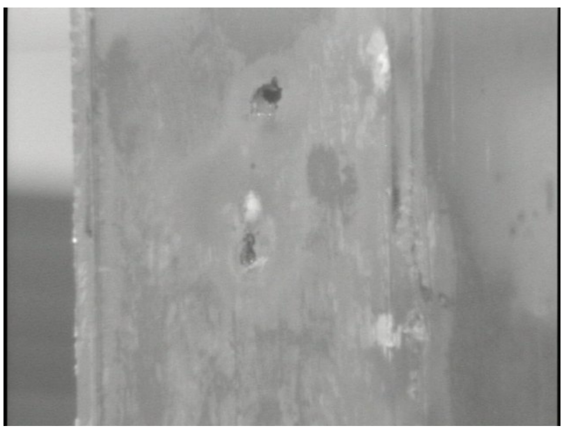

**Figure 2. Pits through the clad on outside fuel plate of fuel Materials Test Reactor equivalent design assembly.**

The containment analysis followed the NRC-approved methodology previously developed at SRS [7]. The methodology was adapted from ANSI 14.5 [8] and NUREG-6487 [9], which provide guidance on the implementation of the regulatory requirements for the shipment of commercial spent nuclear fuel. The bases for the source term components of fines, fission gas, volatiles, and crud as a function of total exposed fuel area and number of breached assemblies were developed based on fuel-specific performance data including laboratory experiments and phenomenological modeling [7].

**Radionuclide Inventory**

A radionuclide production/depletion analysis was performed to estimate the fuel-specific isotopic compositions. Determination of the isotopic content of the spent fuel is made through use of a coupled one-dimensional depletion and shielding analysis module, SAS2h, of SCALE4.4 [10-11]. During exposure, the assembly cladding is assumed intact regardless of assumption used for containment analysis. It is conservatively assumed that the fuel has been decayed for only 10 years. Additional conservative assumptions used in the calculations concerning the development of the radionuclide inventory were derived from Appendix A documents for the RA-3 fuel assemblies [12] and are provided in Table 1.

**Table 1 Input Assumptions for SAS2h**

| Parameter | Assumed Value |
|---|---|
| Specific Power [MW/ass] | 0.06 |
| Burn Time [days] | 1204 |
| Cooling Time [years] | 10 |
| Moderator | $D_2O$ |
| Moderator Temperature [K] | 325 |
| Clad Temperature [K] | 446 |
| Fuel Temperature [K] | 447 |
| Active Meat Thickness [cm] | 0.056 |
| Active Meat Width [cm] | 5.9 |
| Active Meat Length [cm] | 61.5 |
| Number of Plates | 19 |
| Clad Thickness [cm] | 0.042 |
| Pitch [cm] | 0.44 |
| Cladding Material | Aluminum |
| Fuel Material Masses [g/ass] | |
| U-235 | 200.1 |
| U-238 | 22.2 |
| Al | 993.7 |

**Radionuclide Release**

Two cases concerning the degree to which the cladding of each of the RA-3 spent fuel is breached are considered. For the purpose of this analysis, it is assumed that each of the 42 spent RA-3 assemblies per cask exhibit equal amounts of exposed fuel meat surface area.

In the first or "Best-Estimate" case, the surface area of exposed fuel meat of the assemblies is derived from records of the visual inspection of the fuel. Consistent with the fuel storage experience described in Ref. [7], it is assumed that the through clad pitting is limited to the outer fuel plates. The worst case amount of exposed fuel meat per assembly is 3 $cm^3$, based upon records from the visual examination of the fuel [5-6]. Thus 3 $cm^2$/assembly is assumed for each assembly for "normal" transportation conditions and 6 $cm^2$/assembly for the accident conditions of transport, per the methodology in Ref. [7].

For the "Conservative" case, it is assumed that 57 $cm^2$/assembly (i.e., 3 $cm^2$/plate for all 19 fuel plates per assembly) and 114 $cm^2$/assembly (i.e., 6 $cm^2$/plate) for the normal and accident conditions of transport, respectively. The assumed fractional release factors of the various categories of radionuclides transported in a Type B package to the cask free volume are summarized in Table 2. These factors are derived from the containment methodology contained in Ref. [7]. The free volume of the cask was assumed equal to 2.293E+5 $cm^3$ when fully loaded with 42 assemblies.

**Table 2 Data Summary of Inputs to Example Release Calculations**

| Parameter | Best Estimate Normal/Accident | Conservative Estimate Normal/Accident |
|---|---|---|
| **Fraction of Breached Fuel in a Cask, $f_b$** This is the fraction of assemblies in a cask that could release gas, volatiles, and that result in fuel surface area exposure. | 1.0 | 1.0 |
| **Amount of Meat Surface Area Exposed, ESA** | 3 $cm^2$/ass/ 6 $cm^2$/ass | 57 $cm^2$/ass/ 114 $cm^2$/ass |
| **Fission Gas Release Fraction, $f_G$** | 0.30/1.0 | 0.30/1.0 |
| **Volatile Release Fraction, $f_V$** | 1E-06 | 1E-06 |
| **Fraction of Fuel Meat Corrosion Product Layer Released due to Spallation, $T_F$** | 0.15/1.0 | 0.15/1.0 |
| **Crud Spallation Fraction, $f_C$** | 0.15/1.0 | 0.15/1.0 |

**Containment Calculations**

The containment criterion for Type B packages requires that a package have a radioactive release rate less than $A_2 x10^{-6}$ in one hour and $A_2$ per week under normal conditions of transport and for accident conditions, respectively [7]. The parameter $A_2$ has units of curies (Ci) and is isotope dependent. $A_2$ is calculated from the isotopic curie concentration in the fuel as determined through use of the SAS2h module of SCALE4.4.

Input for the analysis was taken from Ref. [7], the Appendix A for the spent fuel [12], and the documentation of the visual inspection [5]. The analysis followed the methodology developed by WSRC for performing containment analyses for breached Al-SNF [7].

Assuming that the release rate is independent of time, the maximum permissible release rates for normal ($R_N$) and accident ($R_A$) conditions of transport, respectively, can be expressed as follows:

$$R_N = L_N C_N \le A_{2,N} \text{ x } 2.78\text{x}10^{-10} \text{ /sec,} \qquad (1)$$
$$R_A = L_A C_A \le A_{2,A} \text{ x } 1.65\text{x}10^{-6} \text{ /sec,} \qquad (2)$$

where:

$R_i$ is the release rate for normal ($R_N$) and accident ($R_A$) conditions of transport [Ci/s],

$L_i$ is the volumetric gas leakage rate [$cm^3$/s] under normal ($L_N$) and accident ($L_A$) conditions of transport,

$C_i$ is the curies per unit volume of the radioactive material, "activity density", that passes through the leak path for normal ($C_N$) and accident ($C_A$) conditions of transport [Ci/cm$^3$], and

$A_{2,i}$ is the group $A_2$ of the radionuclides available for release under normal $A_{2,N}$ and $A_{2,A}$ accident conditions of transport [Ci].

The group $A_2$ values are determined from the isotopic activity concentration in the fuel along with the assumptions listed in Table 2. These calculations also result in the calculation of the activity density terms in Eqs. 1 & 2. Using calculated group $A_2$ values in Eqs. 1 & 2 yields the maximum permissible release rates. Substituting calculated values for activity density and release rate into the same equations yields the volumetric gas leakage rate at the maximum temperature and pressure change across the cask boundary expected during transport. The resulting gas leakage rate is then converted to the maximum leakage rate at standard conditions by making assumptions about the geometry and size of the leak path.

**Leak Path Modeling**

The volumetric gas leak rate is modeled as a combination of continuum and molecular flow through a single leak path. The leak path is modeled as a smooth, right-circular cylinder with sharp edges. To correlate the maximum permissible leak rates to the leak rate at standard temperature and pressure, the leak rate model is implemented assuming the maximum expected internal cask temperature and the maximum expected pressure drop across the leak path (i.e., the cask boundary) to determine the capillary diameter of the leak path. The calculated diameter is then used in the leak rate model at 298-K with internal and external pressures equal to 1.00-atm and 0.01-atm, respectively. The assumptions used in this conversion are provided in the Table 3.

**Table 3 Assumptions Used in Correlation of Maximum Permissible Leakage Rates to Standard Leakage Rates**

| Parameter | Normal Conditions | Accident Conditions |
|---|---|---|
| Backfill Gas | He | He |
| Capillary Length [cm] | 1.0 | 1.0 |
| Gas Temperature [K] | 474 | 574 |
| Upstream Press. [atm] | 1.99 | 11.4 |
| Downstream Press. [atm] | 1.00 | 1.00 |

The calculated standard leakage rates are used to determine whether the loaded cask complies with the containment requirements for shipping spent fuel. These values are compared against the measured cask leakage rate at standard conditions. The measured cask leakage rate must not exceed the calculated maximum leakage rate of the cask, $L_R$, at standard conditions to comply with the containment requirements.

**Containment Analysis Results**

The results of the containment analysis of the RA-3 spent fuel using conservative assumptions are presented in Table 4. These results indicate that the spent fuel can be transported in a standard shipping cask and maintained within the allowable release rates under normal and accident conditions.

The limiting standard leakage rates for the best estimate and the conservative cases correspond to normal conditions of transport. These limiting standard leakage rates indicate that a shipping cask, with a test leakage rate less than $2.89 \times 10^{-5}$ atm·cm$^3$/sec, can safely contain the spent fuel from either case during transport. This result is facilitated by low fuel burnup and long cooling time prior to shipment.

**Table 4 Determination of Maximum Leakage Rates [$L_R$] at Standard Conditions for Shipment of RA-3 Fuel**

| Parameter | Best Estimate | | Conservative Estimate | |
|---|---|---|---|---|
| | ***Normal Transport Conditions*** | ***Accident Transport Conditions*** | ***Normal Transport Conditions*** | ***Accident Transport Conditions*** |
| **Group $A_2$ [Ci]** | 2.19E+02 | 1.56E+02 | 7.80E+1 | 2.55E+1 |
| **R [Ci/s]** | 6.10E-08 | 2.57E-04 | 2.17E-8 | 4.21E-5 |
| **C [Ci/cm$^3$]** | 6.57E-04 | 2.19E-03 | 6.59E-4 | 2.21E-3 |
| **L [cm$^3$/sec]** | 9.28E-05 | 7.17E-01 | 3.29E-5 | 1.90E-2 |
| **$L_R$ [std cm$^3$/sec]** | **8.35E-05** | **1.79E-02** | **2.89E-5** | **2.94E-3** |

## IMPACT OF EXPECTED RADIOACTIVITY RELEASE TO BASIN WATER

The effect of directly storing the breached fuel on the activity of the L-basin at SRS was evaluated using the expected corrosion rate of the exposed fuel meat. This section addresses the potential impact of the introduction of a bounding RA-3 spent fuel assembly on basin activity with respect to the potential release of radioactive $^{137}$Cs into the basin water. Past experience at SRS indicates that monitoring $^{137}$Cs provides a general trend for overall basin water activity. This is due primarily to the fact that the activity in the basin is predominately $^{137}$Cs.

The bounding assembly is a hypothetical construct of an assembly with a burnup of 72.24 MWd (~45%), a cooling time of 10 years, and 3 cm$^2$/assembly of exposed fuel meat material. The burnup is chosen based upon the highest burnup provided in the Appendix A's for the spent fuel [12]. The minimum cooling time provided in the Appendix A's is approximately 12 years, and a conservative value of 10 years has been evaluated. The value for the surface area of exposed fuel meat is derived from the visual inspection of the individual fuel assemblies in Argentina and is consistent with that used in the containment calculations.

The methodology to evaluate the effect of breached fuel on basin activity was previously established in Ref. [13]. The following subsections summarize this methodology and provide the calculations to estimate the effect of the RA-3 fuel on the L-basin steady state activity level.

### Model for Radioactivity Release Rate from Al-SNF into Water

The basin activity is dominated by the concentration of $^{137}$Cs in the basin water as determined by chemical analysis of basin water samples. Therefore, modeling radiation release rates from Al-SNF into the basin water concentrates on the release rate of $^{137}$Cs. In general, the release of radioactivity from Al-SNF with breached cladding into water is dependent on several factors:

- area of exposed fuel
- environment (temperature and quality of the water);
- radioisotope content of fuel (enrichment, burn-up, and decay time);
- fuel meat material (post-irradiation composition and microstructure); and
- clad material

At the low temperatures typical of basin storage (approximately room temperature), corrosion is the primary mechanism whereby species from the fuel core are released into the water. That is, diffusion transport of species from regions in the fuel core to the exposed fuel surface and direct release is not significant.

The radionuclides are assumed to be fully soluble and free to disperse into the water, and not bound in the corrosion product. A simple model to estimate the release from fuel core has been developed by considering general corrosion of the fuel core region directly exposed to the environment [13]. The release model is given by:

$$R = A \times B \times C \quad (3)$$

where:

- R is the $^{137}$Cs release rate [Ci/hr];
- A is the $^{137}$Cs activity density in the fuel meat material at the decay time of interest [Ci/cm$^3$];
- B is the area of fuel exposed to the environment (area of breach) [cm$^2$]; and
- C is the general corrosion rate of the fuel core material in the environment of exposure [cm/hr].

A single bounding fuel assembly contains approximately 159 curies of $^{137}$Cs (taken from containment calculations). The fuel meat volume is derived from Appendix A data [12] and is equal to 386 cm$^3$/assembly. Therefore the $^{137}$Cs activity density in the meat region (A) is 0.412 Ci/cm$^3$. B represents the total area of exposed fuel meat for all 207 RA-3 assemblies. It is determined by using the data presented in Ref. [5], which provides an estimate of the exposed fuel core surface area for each of the 207 RA-3 assemblies. The resulting exposed meat surface area is 140.4 cm$^2$, and C is assumed equal to 5.808E-8 cm/hr (5E-4 cm/yr), consistent with containment calculations. The $^{137}$Cs release rate into the basin from all RA-3 spent fuel assemblies is calculated to be approximately 3.36 µCi/hr.

**Steady-State Release of Cesium-137 into the Basin**

The activity concentration of the basin is directly related to the pumping rate through the deionizers and the release rate of $^{137}$Cs from the existing basin sources and from the RA-3 fuel meat material exposed by the through clad penetration. The long-term steady-state activity concentration in the basin will be from these sources. The increase in steady-state $^{137}$Cs activity due to the addition of all 207 RA-3 spent fuel assemblies is estimated by the following.

$$Q \bullet C = A \bullet V \quad (4)$$

where:

- Q is the flow rate through the system [gal/day],
- C is the steady-state $^{137}$Cs activity of the water [dpm/ml],
- A is the $^{137}$Cs release rate to the water [dpm/ml/day], and
- V is the basin volume [gal].

This equation is used to first determine the steady-state activity release rate to the basin before adding the RA-3 fuel. The value of Q is assumed to be 288,000 gal/day, consistent with the value used in Ref. [13]. The basin volume is 3,500,000 gallons, and the steady-state activity of the basin water is assumed equal to 5.0 dpm/ml. Using the previous equation, it is calculated that the steady-state release rate prior to the addition of the RA-3 fuel assemblies is 0.411 dpm/ml/day. An additional 0.014 dpm/ml/day (3.36 µCi/hr) $^{137}$Cs activity would be released from the RA-3 assemblies. Hence, the total release rate, including existing basin sources and that due to the addition of all 207 RA-3 spent fuel assemblies, is 0.42 dpm/ml/day. This new value (A) is used in the above equation to determine the value of C. The result is that the introduction of the RA-3 assemblies will cause the steady-state activity concentration in the basin to increase from 5.0 dpm/ml to 5.2 dpm/ml compared to the basin operation limit of 200 dpm/ml for $^{137}$Cs activity. These calculations indicate that the addition of the RA-3 spent fuel assemblies will have little impact on the steady-state activity concentration in the basin.

The steady-state activity concentration in the basin would be expected to increase slightly from 5.0 dpm/ml to approximately 5.2 dpm/ml with the addition of the RA-3 fuel using the best-estimate for the maximum exposed fuel area for each of the 207 assemblies. These values are well below the operations limit of 200 dpm/ml.

**Basin Release Calculation Results**

The calculations presented above indicate that the introduction of all 207 RA-3 fuel assemblies, with a combined total exposed meat surface area of 140.4 $cm^2$, into L-Basin would result in a 0.2 dpm/ml increase in the steady-state $^{137}Cs$ activity in the basin, resulting in a steady-state value of 5.2 dpm/ml. This value is well below the basin operations limit of 200 dpm/ml for $^{137}Cs$ activity. Based upon these calculations and assumptions, it is determined that the addition of the RA-3 spent fuel will have negligible impact on safe basin operations.

Using the estimated rate of 5 µm/year for the corrosion rate of both the aluminum cladding and exposed fuel meat in the SRS basin water chemistry, insignificant damage or reconfiguration of the fuel would occur under a period of extended storage (e.g., 20 years). Therefore, the parameters are assumed to be a constant over the storage period.

The storage conditions present in the CSF basins promote the continued corrosion of the aluminum fuel. It is fully expected that the storage of the RA-3 fuel in the SRS L-basin will mitigate the corrosion degradation and that the fuel can be safely stored for up to several decades allowing for acceptable basin storage, retrieval, and ultimate disposition alternatives.

**CONCLUSIONS**

The results of the containment analysis for the RA-3 fuel indicate that the spent fuel can be transported, without canning operations, in a standard shipping cask with a leak rate less than $2.89 \times 10^{-5}$ atm·$cm^3$/sec and maintained within the allowable release rates under normal and accident conditions. It has been further determined that the degraded condition of the fuel requires no canning with respect to basin storage at SRS. The handling and storage operations at SRS will not result in a significant impact on either the condition of the degraded fuel assemblies or on basin operations.

## Acknowledgments

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